Canted ground state in artificial molecules at high magnetic fields.

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We analyze the transitions that a magnetic field provokes in the ground state of an artificial homonuclear diatomic molecule. For that purpose, we have performed numerical diagonalizations for a double quantum dot around the regime of filling factor 2. We present phase diagrams in terms of tunneling and Zeeman couplings, and confinement strength. We identify a series of transitions from ferromagnetic to symmetric states through a set of canted states with antiferromagnetic couping between the two quantum dots.

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It is well known [1], that the ground state (GS) of homonuclear diatomic molecules, as C_2, O_2 or N_2, suffers a series of transitions when the distance between the nuclea is varied. Particularly interesting are the singlet-triplet transitions, which determine the magnetic properties of gases formed with those molecules. Since the intramolecule interatomic distance is, essentially, constant it is not easy to control the physical mechanisms responsible for such a behavior. Recently, double quantum dots (DQD), which can be thought of as artificial homonuclear molecules [2,3], have opened up new possibilities, that should allow both a better understanding of the physics of these systems and the tailoring of their magnetic properties, as some parameters, which are fixed in natural molecules, can be continuously varied in DQD. For instance, it is possible to change potential barrier heights, thus varying the tunneling rate between the two artificial atoms, without altering the interdot distance, which determines the electron-electron interaction. Also, the interdot distance is a parameter that can be externally controlled by appropriate design of the nanostructure defining the DQD.

The use of an external magnetic field B to produce transitions in the GS is a particularly interesting tool to study artificial molecules: apart from the effect of Zeeman energy, the magnetic field introduces an additional length scale (the magnetic length) that, even for moderate fields, can be of the order of the molecular dimensions in these artificial systems. In this work we analyze the symmetric-ferromagnetic transition which occurs in DQD's with N electrons as the magnetic field is increased. Some previous works [4,5] have dealt with DQD in magnetic fields, but in a totally different regime: there, the spin degree of freedom was not included, i.e., those works only considered magnetic fields high enough for the system to be always in the ferromagnetic phase.

We restrict ourselves to *homopolar* molecules, i.e., DQD with N even where the electrons are three dimensionally confined: in the z direction they can reside in either one of two layers (top (T) or bottom(B)), separated by a distance d, while in the $x-y$ plane electrons are confined by the presence of a two-dimensional parabolic potential $V(\vec{r}) = m^*\omega_0 r^2/2$ (m^* is the electron mass, r is the in-plane distance) and to a range of magnetic fields such that the DQD has a filling factor close to $\nu = 2$. This last restriction is motivated by the symmetric-ferromagnetic transitions found in infinite multicomponent Quantum Hall (QH) systems close to filling factor $\nu = 2$ [6].

We work in the symmetric gauge, with a set of states restricted to the lowest orbital Landau level, i.e. single particle wavefunctions without nodes in the radial direction. In this basis set, the Hamiltonian of the DQD is

$$H = \alpha M - \Delta_z S_z - \frac{\Delta_{sas}}{2}\sum_{m\sigma}\left(c^\dagger_{m\sigma T}c_{m\sigma B} + c^\dagger_{m\sigma B}c_{m\sigma T}\right) + \sum_{\{m_j\}\sigma\sigma'\Lambda\Lambda'}\frac{V^{\Lambda\Lambda'}_{m_1m_2m_3m_4}}{2}c^\dagger_{m_1\sigma\Lambda}c^\dagger_{m_2\sigma'\Lambda'}c_{m_3\sigma'\Lambda'}c_{m_4\sigma\Lambda}, \quad (1)$$

where $\omega_c = eB/m^*$ and $\alpha = \hbar([\omega_c^2 + 4\omega_0^2]^{1/2} - \omega_c)/2$. m and M are the third components of single particle and total angular momentum respectively. σ and S_z are the third components of single particle and total spins respectively and Λ a layer index, equal to T or B. $\Delta_z = g\mu_B B$ is the Zeeman coupling with g being the Landé g-factor and μ_B the Bohr magneton. Δ_{sas} is the single particle energy gap between symmetric (s) and antisymmetric (as) combinations of T and B single particle states. $V^{\Lambda\Lambda}$=$e^2/(\varepsilon r)$ and $V^{\Lambda\Lambda'}$=$e^2/(\varepsilon\sqrt{r^2+d^2})$ for $\Lambda \neq \Lambda'$ are Coulomb interaction potentials, e being the electron charge and ε the dielectric constant. In all our results, the energies are given in units of $e^2/(\varepsilon l_B)$ with

$l_B = [\hbar/(m^*[\omega_c^2 + 4\omega_0^2]^{1/2})]^{1/2}$ being the magnetic length. The eigenstates of the DQD are a function d, Δ_z, Δ_{sas} and α. The spectrum is separated in subspaces labeled by the quantum numbers (M, S_z, P_I) where $P_I = (-1)^{I/2}$ is the parity of the isospin $I = N_s - N_{as}$ with N_s, N_{as} being the number of electrons in symmetric and antisymmetric states respectively. It should be noted that, for $d \neq 0$, interdot and intradot Coulomb interactions are different. In this case, electron-electron interaction mixes states with different isospin, and I is not a good quantum number. However, the parity P_I is always a good quantum number because it is related to a symmetry operation of the problem: the reflection $\mathcal{R}$ with respect to the midplane between the two layers. The application of $\mathcal{R}$ to any state (not necessarily an eigenstate) with a well defined isospin would result in the same state multiplied by $(-1)^{N_{as}} = (-1)^{(N-I)/2}$ which, apart from the unessential constant $(-1)^{N/2}$, is precisely P_I times the state. This shows that $\mathcal{R}$ applied to any state without a well defined isospin would still produce the same state (except for a sign), provided the initial state had weight in different isospin subspaces with the same isospin parity. There is another symmetry operation in the problem: the inversion with respect to the midpoint between the centers of the two quantum dots (QD). We do not pay special attention to it in the discussion of our results, because it does not add any information of relevance.

We have diagonalized numerically the Hamiltonian (1) for $N = 6$ and $N = 8$. As similar structure is found in both cases, only results for $N = 8$ are presented here.

The analysis of the results will be helped by the large experience accumulated in the knowledge of the GS of a multicomponent QH systems [6]. When electrons are confined in a double layer (DL) in the regime of global filling factor $\nu = 2$, they have as degrees of freedom σ ($\equiv\uparrow, \downarrow$) and layer index Λ. Those degrees of freedom provoke a rich phase diagram in terms of tunneling and Zeeman couplings. Obviously, when Zeeman splitting Δ_z is much larger than tunneling splitting Δ_{sas}, the system prefers a ferromagnetic GS $|F\rangle$ in which electrons occupy all the symmetric and antisymmetric states with spin $\uparrow$. In the opposite limit $\Delta_{sas} >> \Delta_z$, the GS $|S\rangle$ corresponds to electrons fully occupying the symmetric states with both $\uparrow$ and $\downarrow$ spins. The character of the transition between these two extreme situations is controlled by electron-electron interaction effects in the four-dimensional space of degrees of freedom. A very attractive proposal was made [7–9] for a GS, labeled as canted, which connects continuously between the $|F\rangle$ and $|S\rangle$ limiting cases. The canted state is a ferromagnet in the field direction while, for the direction perpendicular to the magnetic field, spins in different layers have antiferromagnetic correlations. The properties of this state has been studied by a microscopic Hartree-Fock theory, a long wavelength field theory based on the quantum O(3) nonlinear sigma model and a bosonic spin theory [7–14]. Moreover, exact numerical diagonalizations for a small number of electrons in a DL with spherical shape [15] show that, in a translationally invariant system, the canted phase survive quantum fluctuations; although the domain, in the (Δ_z, Δ_{SAS}) parameter space, in which it is the GS is much narrower than what Hartree-Fock approximation predicts. From the experimental side, such a phase is consistent with the available information in DL by inelastic light scattering [16], magnetoresistance [17] and capacitance spectroscopy [18].

In our calculations in DQD, we also find that, for large Δ_z and moderate Δ_{sas}, the DQD has a $|F\rangle$ GS while for large Δ_{sas} and moderate Δ_z the GS is $|S\rangle$. As in the infinite system, the DQD presents a transition between these two regimes through a set of more complicated states.

In order to identify a GS obtained from numerical diagonalizations as a canted state, a difficulty arises from the fact that the eigenstates of the DQD have a well defined third component S_z of the spin while the mean field wave function $|C^{MF}\rangle$ for the canted state is not an eigenstate of S_z. However, we can restore the broken symmetry of the mean field states by projecting on subspaces with well defined $S_z = N/2 - n$:

$$|C_n^{MF}\rangle = \int d\phi e^{i\phi n} |C^{MF}(\phi)\rangle \tag{2}$$

where n is an integer number. In Eq. (2), ϕ is the angle defining a particular canted state $|C^{MF}(\phi)\rangle = (\Phi_{1-}(\phi), \Phi_{2-}(\phi))$ in which, for filling factor 2, the electrons are occupying two type of states

$$\Phi_{1-}(\phi) = \begin{pmatrix} cos(\theta_1/2) \\ -e^{i\phi} sin(\theta_1/2) \\ 0 \\ 0 \end{pmatrix} ; \Phi_{2-}(\phi) = \begin{pmatrix} 0 \\ 0 \\ e^{i\phi} cos(\theta_2/2) \\ -sin(\theta_2/2) \end{pmatrix}. \tag{3}$$

The notation is the one used by Das Sarma *et al.* [9] with the states (3) given in the basis of $(s, \uparrow)$, $(as, \downarrow)$, $(s, \downarrow)$, $(as, \uparrow)$. The angles θ_i depend on the Hamiltonian parameters being $\theta_1 = \theta_2 = 0$ for the ferromagnetic state $|F\rangle$ and $\theta_1 = 0$ and $\theta_2 = \pi$ for the symmetric state $|S\rangle$. After some algebra, one gets the projections (2) of the canted state written as

$$|C_n^{MF}\rangle = \mathcal{C}_n \left[cos(\theta_1/2) cos(\theta_2/2) \sum_m c^\dagger_{m,as,\downarrow} c_{m,s,\uparrow} + sin(\theta_1/2) sin(\theta_2/2) \sum_m c^\dagger_{m,s,\downarrow} c_{m,as,\uparrow} \right]^n |F\rangle \tag{4}$$

$\mathcal{C}_n$ being a normalization constant. The coefficients in Eq.(4) are m-independent in the infinite system because all single particle states are degenerate in the lowest Landau level. However, in a parabolic QD, translational invariance is broken, the single particle energy depends on m and the corresponding canted eigenstates of the DQD

should have a structure similar to Eq.(4) although the coefficients θ_1^m and θ_2^m could have a dependence on m.

Fig. 1 contains our results in a phase diagram showing the quantum numbers (M, S_z, P_I) of the GS for $d = l_B$ and $N = 8$. In order to work in the regime $\nu = 2$, we chose $\alpha = 0.2e^2/(\varepsilon l_B)$. In the region to the left corresponding to small Δ_{sas}, the GS, labeled as (12,4,1) is a ferromagnetic state given by a single Slater determinant $|F\rangle = \prod_{m=0}^{3} c^\dagger_{m,s,\uparrow} c^\dagger_{m,as,\uparrow}$. There is another large ferromagnetic region labeled as (13,4,-1) with a GS which is practically (more than 98%) one Slater determinant $|F'\rangle = \prod_{m=0}^{4} c^\dagger_{m,s,\uparrow} \prod_{m=0}^{2} c^\dagger_{m,as,\uparrow}$. This means that for an increasing tunneling, the ferromagnetic state $|F\rangle$ suffers an edge reconstruction by a charge instability which preserves the ferromagnetic character giving $|F'\rangle$. In the lower region corresponding to small Δ_z, the GS, labeled as (12,0,1), is a symmetric state given by practically (more than 98%) one Slater determinant $|S\rangle = \prod_{m=0}^{3} c^\dagger_{m,s,\uparrow} c^\dagger_{m,s,\downarrow}$. When the Zeeman coupling increases, there is a new symmetric GS, labeled as (13,1,1), which is also practically given by $|S'\rangle = \prod_{m=0}^{4} c^\dagger_{m,s,\uparrow} \prod_{m=0}^{2} c^\dagger_{m,s,\downarrow}$. The increase of Δ_z provokes an edge reconstruction which involves a charge-spin excitation giving a new GS which essentially preserves the symmetric character.

The most interesting part of the phase diagram corresponds to the narrow regions separating ferro-like regions from symmetric-like regions. The regions (12,3,-1), (12,2,1) and (12,1,-1) contain canted states $|C_n\rangle$ (corresponding to $n = 1, 2, 3$ respectively) while the regions (13,3,1) and (13,2,-1) contain canted states $|C'_n\rangle$ (corresponding to $n = 1, 2$ respectively). This identification is made through the wavefunctions which turn out to have the functional form given by Eq. (4) with coefficients θ_1^m and θ_2^m almost independent on m at the center of each QD. Since these regions are well described by states (4), they must be understood as the projection to well defined quantum numbers of canted states with an antiferromagnetic tilting of the spins.

Apart from the states shown in Fig. 1, some other GS, in particular with different edge reconstructions, could appear when the parameters are varied in the calculation. In fact this is the case: as shown in figures 2 and 3, showing phase diagrams for $d = l_B$, $N = 8$, when Δ_z and α are varied for fixed $\Delta_{sas} = 0.1e^2/(\varepsilon l_B)$ and $\Delta_{sas} = 0.2e^2/(\varepsilon l_B)$ respectively. In the central part of Fig. 2, the sequence (12,4,1), (12,3,-1), (12,2,1), (12,1,-1) and (12,0,1) corresponds to the already discussed transition $|F\rangle \to |C_n\rangle \to |S\rangle$ with $n = 1, 2, 3$. Contiguously to its left, for smaller values of α, the sequence (13,4,-1), (13,3,1), (13,2,-1) and (13,1,1) corresponds to the transition $|F'\rangle \to |C'_n\rangle \to |S'\rangle$ with $n = 1, 2$. To the right of those regions, the increase in α implies a higher confinement and, consequently, a larger concentration of the electrons in the center of the DQD which, in turns, implies a decrease of M. Edge reconstruction occurs in the leftmost region, i.e., smallest α, where a reduction of the confinement provokes the spreading of the electron density away from the center. In Fig. 3 the tunneling is so large that the sequence (12,4,1), (12,3,-1), (12,2,1) and (12,1,-1) does not appear and only the symmetric state $|S\rangle \equiv (12, 0, 1)$ is observed. Apart from this, the whole structure is similar to that shown in Fig. 2.

In summary, we have analyzed the transitions that a magnetic field provokes in the GS of an artificial homonuclear diatomic molecule. For that purpose, we have carried out numerical diagonalizations for a DQD for filling factors close to $\nu = 2$. The resulting phase diagrams, are understood to the light of previous experience on the GS of an infinite DL at the same regime. When the different parameters are varied, a series of transitions $|F\rangle \to |C_n\rangle \to |S\rangle$ from ferromagnetic to symmetric states are identified, through a set of projections (into subspaces with well defined quantum numbers) of canted states. Such canted states have ferromagnetic correlations in parallel (to magnetic field) direction and antiferromagnetic correlations in the perpendicular components of spins in different QD. The electron-electron correlation that leads to the canted states in infinite double layers for $\nu = 2$ survives, in this regime, to the presence of edges and is present even when there is edge reconstruction.

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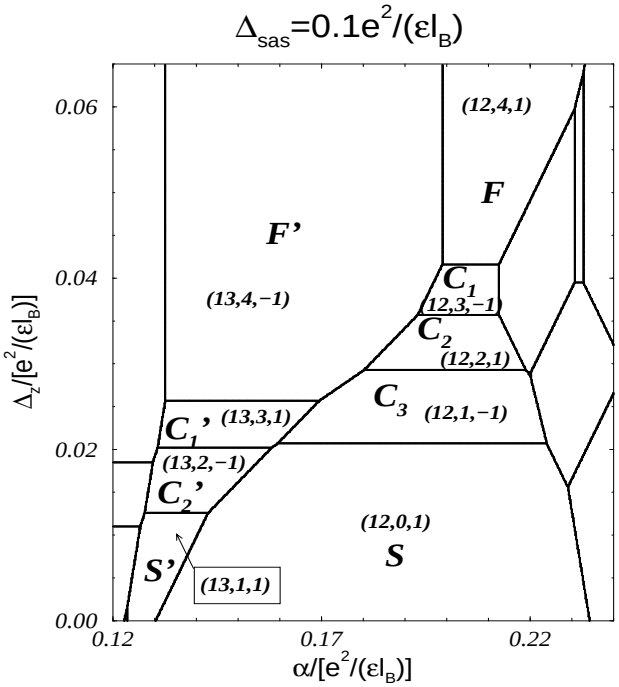

FIG. 2. Phase diagram showing the subspace (M, S_z, P_I) containing the GS for $d = l_B$, $\Delta_{sas} = 0.1e^2/(\varepsilon l_B)$ and $N = 8$. Energies α and Δ_z are given in units of $e^2/(\varepsilon l_B)$.

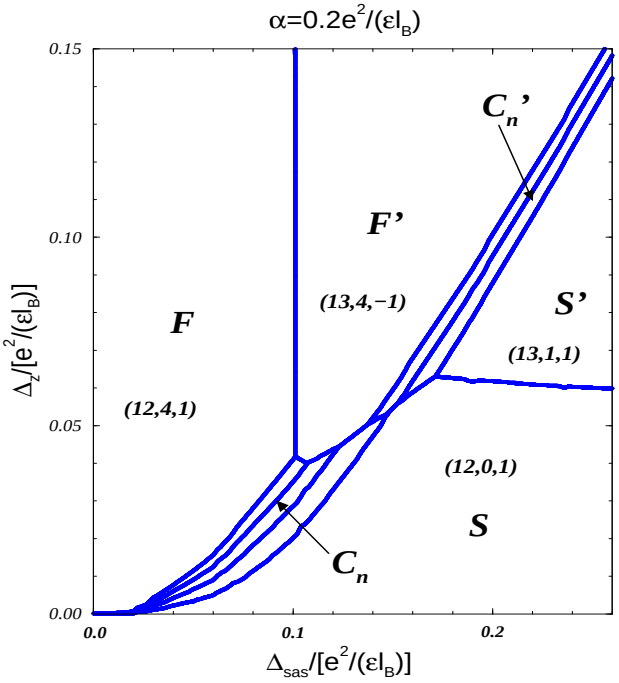

FIG. 1. Phase diagram showing the subspace (M, S_z, P_I) containing the GS for $d = l_B$, $\alpha = 0.2e^2/(\varepsilon l_B)$ and $N = 8$. Energies Δ_{sas} and Δ_z are given in units of $e^2/(\varepsilon l_B)$.

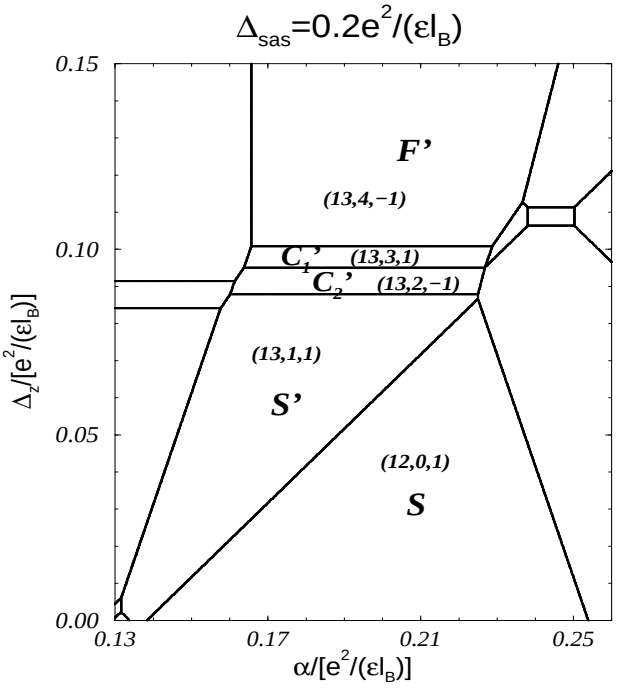

FIG. 3. Phase diagram showing the subspace (M, S_z, P_I) containing the GS for $d = l_B$, $\Delta_{sas} = 0.2e^2/(\varepsilon l_B)$ and $N = 8$. Energies α and Δ_z are given in units of $e^2/(\varepsilon l_B)$.